

PATENT
Case 4233C3

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: Donald B. Appleby et al : Group Art Unit: 1211
Serial No.: 08/360,184 : Examiner: E. White
Filed: December 20, 1994 :
For: **Polyol Polyester Synthesis**

FIRST DECLARATION UNDER 37 C.F.R. §1.608 OF SCOTT D. PEARSON

Assistant Commissioner for Patents
Washington, DC 20231

Dear Sir:

I, SCOTT D. PEARSON, declare that:

1. I am a co-inventor of and am familiar with the present U.S. patent application.
2. I received a Bachelor of Chemical Engineering Degree from the University of Minnesota in 1981 and a Doctorate of Philosophy in Chemical Engineering from Iowa State University in 1987.
3. I was employed by the assignee of the present application, The Procter & Gamble Company, from about February 1987 to about February 1991. During my employment with The

Procter & Gamble Company, I worked, inter alia, on developing a continuous process for the synthesis of polyol fatty acid esters, specifically sucrose fatty acid esters, referred to herein as sucrose polyesters, by reaction of sucrose and fatty acid methyl esters.

4. From about June 1988 to about July or August 1990, I worked as a Group Leader on the continuous sucrose polyester production project. My responsibilities included coordinating and supervising pilot plant runs of the continuous sucrose polyester production process, analyzing data collected during the pilot plant runs, and preparing reports summarizing the individual pilot plant runs, results thereof and the progress of the operation of the continuous sucrose polyester production equipment.

5. From January 18, 1989 to January 27, 1989, a pilot plant run of the continuous sucrose polyester production process was performed under my direction and control using the pilot plant located in Building 96 of The Procter & Gamble Company's Ivorydale plant. The January 18, 1989 to January 27, 1989 pilot plant run was designated as the P90117 pilot plant continuous process. During pilot plant runs of the continuous sucrose polyester production process, including during the P90117 pilot plant continuous process, the operation of the continuous sucrose polyester production equipment, sample collection, analysis of collected samples, and data recordation and review were performed under my direction and control in accordance with established procedures.

6. I have reviewed the respective Declarations Under 37 C.F.R. §1.608 of Pamela D. Hardy, Gene P. Hawkins, David R. Loggains, Michael W. McIntosh, Harold R. McPeek, Milton C. McMullen, Carol L. White and K. Michael Wieser and Exhibits Nos. 1, 3-5, 7-9 and 27 identified in those Declarations, all of which Declarations and Exhibits are submitted herewith. The Declarations of Pamela D. Hardy, Gene P. Hawkins, David R. Loggains, Michael W. McIntosh, Harold R. McPeek, Milton C. McMullen, Carol L. White and K. Michael Wieser accurately describe the procedures used to prepare feed batches for the P90117 pilot plant continuous process, the general reactor set up employed in the P90117 pilot plant continuous process, the procedures used to analyze samples from reactor recirculation loops during the P90117 pilot plant continuous process and the procedures for recording data during the P90117 pilot plant continuous process, as respectively described therein.

7. As set forth in the McIntosh Declaration, Exhibit 1 comprises an accurate copy of page 4.1 of the "Continuous Pilot Plant Start-Up Manual" dated September 26, 1986. Exhibit 1 comprises a general schematic diagram of the reactors employed in the pilot plant equipment used in the P90117 pilot plant continuous process, with various modifications having been made to the pumps, valves and recirculation loops shown in Exhibit 1 for the P90117 pilot plant continuous process. Specifically, the pilot plant equipment employed in the P90117 pilot plant continuous process comprised a catalyst batch reactor 501 from which a catalyst batch was supplied and a series of seven 30-gallon continuous stirred tank reactors (CSTRs), R600-R606, respectively, in which the sucrose polyester reaction was conducted. Flow to each reactor comprised raw material feed and/or flow controlled reactor product from the previous reactor,

and discharge from each reactor was by level control. More particularly, and as described in detail below in paragraph 10, a sucrose feed batch and a catalyst batch were fed to the first reactor R600. Catalyst batch was also fed to the second through fourth reactors, R601-R603, respectively, while additional fatty acid methyl esters were fed to the second and third reactors, R601 and R602, respectively, from a reactor 008 or from a tote. All of the reactors were operated under vacuum and reactors R603-R606 employed nitrogen sparging. Throughout the P90117 pilot plant continuous process, samples were taken from the recirculation loops of the reactors R600 -R606, the samples being representative of the respective reactor's out-going product stream.

8. I have reviewed Exhibit 2 and confirm that it is an accurate copy of the P90117 Experimental Test Plan dated January 12, 1989, which I prepared on or about January 12, 1989. The P90117 Experimental Test Plan sets forth the conditions under which the P90117 pilot plant continuous process was to be run and the conditions under which the P90117 pilot plant continuous process was run from January 18-January 22, 1989. As set forth on page 2 of Exhibit 2, the P90117 pilot plant continuous process employed the seven CSTRs, R600-R606, respectively, in series. The first reactor, reactor R600, comprised an initial reaction stage and the second through seventh reactors, R601-R606, respectively, comprised subsequent reaction stages.

9. In the P90117 pilot plant continuous process, polyol fatty acid esters (sucrose fatty acid polyesters) were synthesized by reacting a polyol (sucrose) and a fatty acid lower alkyl

ester (fatty acid methyl ester) under substantially solvent free conditions in the presence of a transesterification catalyst (potassium carbonate) and an emulsifier (potassium soap). As evidenced in the following paragraphs, the P90117 pilot plant continuous process comprised an initial reaction stage carried out in the first reaction zone (reactor R600) under such conditions that the reaction mixture in the first reaction zone (reactor R600) was in steady state, with overall mass balanced ingoing reactant streams and outgoing product streams. The ingoing reactant streams included, among other components, polyol (sucrose) and fatty acid lower alkyl ester (fatty acid methyl ester) and the outgoing product streams included, among other components, reaction mixture having a polyol conversion (sucrose conversion) of 1% or more and lower alkyl alcohol (methanol) formed during the initial stage of the synthesis. The process further comprised subsequent reaction stages (reactors R601-R606) in which the reaction mixture from the first reactor R600 was further reacted to the polyol fatty acid esters (sucrose fatty acid esters) in subsequent reaction zones. The polyol fatty acid ester synthesis was the dominating reaction occurring in the first reactor R600 and the only significant reaction in which the polyol (sucrose) and fatty acid lower alkyl ester (fatty acid methyl ester) participated in the first reactor R600 in the P90117 pilot plant continuous process.

10. As described at pages 2-4 of Exhibit 2, a sucrose feed batch comprising sucrose, fatty acid methyl ester and potassium soap emulsifier, and a catalyst batch comprising potassium carbonate catalyst and fatty acid methyl ester were the in-going feed streams to the first reactor R600 in the P90117 pilot plant continuous process. As described in the Wieser Declaration, Exhibits 3 and 4 comprise accurate copies of the control records evidencing the components

employed in forming the sucrose feed batch and the catalyst batch, respectively; for the P90117 pilot plant continuous process. Under my direction and control, and Mr. Wieser's supervision and control, potassium soap was prepared in reactor 001 by the reaction of fatty acid ester (I-1) and potassium hydroxide in the presence of methanol. To prepare the sucrose feed batch, the methanol was evaporated off, and fatty acid methyl ester (methyl ester blend) and sucrose were added to the remaining potassium soap. Under my further direction and control and Mr. Wieser's supervision and control, potassium carbonate and fatty acid methyl ester (ester blend Y) were mixed in reactor 501 to form the catalyst batch. The sucrose feed batch components and the catalyst batch components as described in Exhibits 3 and 4 are set forth in Tables 1 and 2, respectively:

Table 1: Sucrose Feed Batch Components

Component	Pounds per batch
KOH*	115
Fatty Acid Ester (I-1)*	600
Methanol**	1200
Fatty Acid Methyl Ester	3700
Sucrose	800

* The fatty acid ester (I-1) and KOH were reacted prior to addition of methyl ester and sucrose to form the potassium soap emulsifier.

** The methanol was evaporated off prior to the sucrose addition.

Table 2: Catalyst Batch Components

Component	Pounds per batch
Potassium Carbonate	250
Fatty Acid Methyl Ester	750

As both the sucrose feed batch and the catalyst batch were substantially solvent free, the polyol fatty acid polyester synthesis in the first reactor R600 was conducted under substantially solvent free conditions.

11. As further described at pages 2-3 of Exhibit 2, during the P90117 pilot plant continuous process, the sucrose feed batch and the catalyst batch were fed to the first reactor (R600). At start up of the process, outgoing liquid product stream from the first reactor (R600) was fed to a drum for about one hour, and thereafter was fed into the second reactor (R601), the outgoing liquid product stream from the second reactor (R601) was fed into a drum for about one hour, and thereafter was fed into the third reactor (R602), and so forth, until all reactors were full and feeding forward. After start-up, and during the period of time from 2:00 a.m. to 13:00 (1:00 p.m.) on January 20, 1989, all reactors in the P90117 pilot plant continuous process were full and feeding forward.

12. Under my direction and control and Ms. White's supervision and control, technicians monitoring the various operating parameters of the pilot plant during the P90117 pilot plant continuous process observed and accurately recorded various operating parameters on log sheets maintained in the pilot plant for the purpose of recording such parameters on an hourly basis in accordance with established procedures. As set forth in the White Declaration, Exhibit 5 comprises accurate copies of such log sheets on which operating parameters of the P90117 pilot plant continuous process were accurately recorded on an hourly basis. More specifically, on January 20, 1989, for the time period of from 2:00 a.m. to 13:00 (1:00 p.m.), the operating

parameters comprising reactor pressure, reactor temperature, slurry feed batch flowrate, confirmation of operation of catalyst injector and injection cycle (dump time--the period between successive catalyst injections) for reactor R600 as set forth in Exhibit 5 were as follows:

Table 3: Operating Parameters for Reactor R600 on January 20, 1989

Time	Reactor Pressure (mm/Hg)	Reactor Temperature (°F)	Slurry Feed Batch Flowrate (pounds/hour)	Catalyst Batch Operating, 0.136 Pounds/ Injection	Injection Cycle (min.)
2:00 a.m.	15.1	275.1	55.2	yes	11
3:00 a.m.	14.9	274.9	55.0	yes	11
4:00 a.m.	15.3	275.3	54.7	yes	11
5:00 a.m.	15.2	275.1	54.9	yes	11
6:00 a.m.	15.0	275.0	54.9	yes	11
7:00 a.m.	14.9	275.1	54.8	yes	11
8:00 a.m.	15.2	275.7	54.9	yes	11
9:00 a.m.	15.4	275.1	54	yes	11
10:00 a.m.	15.1	275.4	55.3	yes	11
11:00 a.m.	15.1	275.1	55	yes	11
12:00 p.m.	14.5	275.4	55	yes	11
13:00 (1:00) p.m.	14.9	275.3	55.2	yes	11

Exhibit 5 demonstrates that the reactor pressure, reactor temperature, slurry feed batch flowrate, and catalyst batch injection rate (the rate at which the catalyst feed mixture was injected) for the initial reactor, reactor R600, were substantially constant over the time period of from 2:00 a.m. to 13:00 (1:00 p.m.) on January 20, 1989. Particularly, Exhibit 5 demonstrates that there was a continuous introduction of reactants comprising polyol (sucrose) and fatty acid lower alkyl ester (fatty acid methyl ester) into the initial reaction stage at a constant rate.

13. I have reviewed Exhibit 6 and confirm that Exhibit 6 comprises an accurate copy of the Process Log Notebook for the P90117 pilot plant continuous process for the days of January 17, 1989 through January 20, 1989, in which various miscellaneous operating information and observations were recorded by engineers and technicians under my direction and control and Ms. White's supervision and control during the pilot plant process, in accordance with established procedures. As set forth at page 3 of Exhibit 6, under the time period designated "11:00-7:00, 1-18-89", which represented the time period from 11:00 pm, January 17, 1989 to 7:00 am, January 18, 1989, the first reactor R600 began filling at 7:00 a.m. on January 18, 1989 and the level on the first reactor R600 was set at 8". Accordingly, once the first reactor R600 was filled, the contents of the reactor were maintained at a constant level throughout the reaction by level controlled discharge of product from the reactor. Since there was a continuous introduction of reactants into the initial reaction stage in the first reactor R600 at a constant rate during the time period of from 2:00 a.m. to 13:00 (1:00 p.m.) on January 20, 1989, and since the first reactor R600 was not accumulating mass owing to the level control discharge, there was also a continuous removal of product from the initial reaction stage, i.e., the first reactor R600, over the same time period. Thus, the reaction mixture in the first reaction stage, i.e., the first reactor R600, was overall mass-balanced during the time period of from 2:00 a.m. to 13:00 (1:00 p.m.) on January 20, 1989.

14. As set forth in the Loggains, McPeck and Wieser Declarations submitted herewith, Exhibit 7 comprises accurate copies of data sheets which set forth determined weight percentages of potassium carbonate catalyst (as carbonate) and potassium soap (as soap) in

samples from the recirculation loop of the first reactor R600. More specifically, at 2:00 a.m., 6:00 a.m., 9:00 a.m., and 13:00 (1:00 p.m.) on January 20, 1989, the weight percentages of potassium carbonate catalyst (as carbonate) and potassium soap (as soap) in the R600 recirculation loop samples as set forth in Exhibit 7 were as follows:

**Table 4: Weight Percentages of Carbonate and Soap
in R600 Samples on January 20, 1989**

Time	% Carbonate	% Soap
2:00 a.m.	0.24	12.41
6:00 a.m.	0.13	12.68
9:00 a.m.	0.18	12.69
13:00 (1:00 p.m.)	0.19	12.65

As the samples from the recirculation loop are representative of the out-going product stream, Exhibit 7 demonstrates that the weight percentages of catalyst (as carbonate) and potassium soap (as soap) in the out-going stream from the reactor R600 were at substantially constant levels during the P90117 pilot plant continuous process on January 20, 1989 for the time period of from 2:00 a.m. to 13:00 (1:00 p.m.).

15. As set forth in the Hardy and Hawkins Declarations submitted herewith, Exhibit 8 comprises accurate copies of data sheets which set forth determined area percentages of sucrose monoester, diester, triester, tetraester, pentaester, hexaester, heptaester and octaester, respectively, in the sucrose ester in samples from the recirculation loop of the first reactor, reactor R600. More specifically, at 4:00 a.m., 7:00 a.m., 10:00 a.m. and 13:00 (1:00 p.m.) on

January 20, 1989, the respective ester area percentages in the sucrose ester in the R600 recirculation loop samples as set forth in Exhibit 8 were as follows:

**Table 5: Area Percentages of Respective Esters in
Sucrose Ester in R600 Samples on January 20, 1989**

Time	Monoester	Diester	Triester	Tetraester	Pentaester	Hexaester	Heptaester	Octaester
4:00 a.m.	10.4	26.3	35.3	22.1	5.9	0	0	0
7:00 a.m.	10.4	27.8	36.6	22.1	3.2	0	0	0
10:00 a.m.	8.2	27.5	35.6	22.5	6.2	0	0	0
13:00 (1:00 p.m.)	9.9	27.1	36.6	20.9	5.5	0	0	0

As the samples from the recirculation loop are representative of the out-going product stream, Exhibit 8 demonstrates that the respective area percentages, and therefore the respective amounts, of sucrose monoester, diester, triester, tetraester, pentaester, hexaester, heptaester and octaester in the sucrose ester in the out-going stream from the reactor R600 were at substantially constant levels during the P90117 pilot plant continuous process on January 20, 1989 for the time period of from 2:00 a.m. to 13:00 (1:00 p.m.).

16. As further set forth in the Hardy and Hawkins Declarations submitted herewith, Exhibit 8 comprises accurate copies of data sheets which set forth determined I-bar values of the sucrose ester in samples from the recirculation loop of the first reactor R600. The I-bar value represents an average degree of esterification of the sucrose ester based on mole fractions. More specifically, at 4:00 a.m., 7:00 a.m., 10:00 a.m. and 13:00 (1:00 p.m.) on January 20, 1989, the I-bar values as set forth in Exhibit 8 were as follows:

**Table 6: I-Bar Degree of Esterification
for R600 Samples on January 20, 1989**

Time	I-Bar Degree of Esterification
4:00 a.m.	2.57
7:00 a.m.	2.53
10:00 a.m.	2.63
13:00 (1:00 p.m.)	2.56

As the samples from the recirculation loop are representative of the out-going product stream, Exhibit 8 demonstrates that the I-bar values of the sucrose ester in the out-going stream from reactor R600 were substantially constant during the P90117 pilot plant continuous process on January 20, 1989 for the time period of from 2:00 a.m. to 13:00 (1:00 p.m.), indicating that a substantially constant degree of esterification was obtained in reactor R600 over the same time period. The I-bar degree of esterification for the sucrose esters in each of the reactor samples was in excess of 2, indicating that, on average, in excess of 2 of the 8 esterifiable groups per sucrose molecule had been esterified, whereby the degree of esterification of the sucrose ester in the out-going product stream from the first reaction stage, reactor R600, was in excess of 25%.

17. As set forth in the Loggains, McPeck and McMullen Declarations submitted herewith, Exhibit 9 comprises accurate copies of data sheets which set forth determined weight percentages of unreacted sucrose in samples obtained from the recirculation loop of the first reactor R600. More specifically, at 2:00 a.m., 6:00 a.m., 7:00 a.m., 8:00 a.m., 9:00 a.m. and 13:00 (1:00 p.m.) on January 20, 1989, the weight percentage of unreacted sucrose in the R600 recirculation loop samples as set forth in Exhibit 9 were as follows:

**Table 7: Weight Percentage of Unreacted Sucrose
in R600 Samples on January 20, 1989**

Time	% Sucrose
2:00 a.m.	1.4
6:00 a.m.	1.03
7:00 a.m.	1.47
8:00 a.m.	1.55
9:00 a.m.	1.33
13:00 (1:00 p.m.)	1.39

As the samples from the recirculation loop are representative of the out-going product stream, Exhibit 9 demonstrates that the weight percentage of unreacted sucrose in the out-going stream from the first reactor R600 was at a substantially constant level during the P90117 pilot plant continuous process on January 20, 1989 for the time period of from 2:00 a.m. to 13:00 (1:00 p.m.).

18. As discussed in ¶12 and ¶17, respectively, the data set forth in Exhibit 5 demonstrates that for the time period of 2:00 a.m. to 13:00 (1:00 p.m.) on January 20, 1989, incoming sucrose to the first reactor R600 (i.e. the sucrose feed batch flow to reactor R600) was at a substantially constant rate and the data set forth in Exhibit 9 demonstrates that the weight percentage of out-going unreacted sucrose leaving reactor R600 was substantially constant. As established in paragraph 13, the first reactor R600 was mass balanced and not accumulating mass, and as set forth in paragraph 9, the sucrose polyester synthesis was the only significant reaction in which the sucrose and fatty acid methyl ester participated in the first reactor R600. Thus, the amount of reacted sucrose, i.e., in the form of sucrose esters, in the out-going product

stream leaving the first reactor R600 was therefore also substantially constant for the same time period.

19. As discussed above in ¶¶15-16, the data set forth in Exhibit 8 indicates that for the period of 2:00 a.m. to 13:00 (1:00 p.m.) on January 20, 1989, the area percentage distribution of the respective sucrose esters in the sucrose ester product and the I-bar average degree of esterification of the sucrose ester product in the out-going product steam leaving the first reactor R600 were substantially constant. Since the amount of sucrose esters in the out-going product stream leaving the first reactor R600 was also substantially constant for the same time period (¶18) and the sucrose-fatty acid methyl ester reaction to form sucrose esters was the only significant reaction occurring in reactor R600 (¶9), the amounts of the respective individual sucrose esters and unreacted fatty acid methyl ester in the out-going product stream leaving reactor R600 were also substantially constant for the same time period.

20. During the transesterification reaction, sucrose was reacted with fatty acid methyl ester to form sucrose esters and methanol. Since the amounts of the respective sucrose esters in the out-going product stream leaving reactor R600 were substantially constant for the time period of 2:00 p.m. to 13:00 (1:00 p.m.) on January 20, 1989, and reactor R600 was not accumulating mass, the amount of methanol leaving reactor R600 was also substantially constant for the same time period.

21. To summarize, during the P90117 pilot plant continuous process on January 20, 1989, for the time period of from 2:00 a.m. to 13:00 (1:00 p.m.), polyol fatty acid esters were synthesized by reacting sucrose and fatty acid methyl ester under substantially solvent free conditions in the presence of a transesterification catalyst, namely potassium carbonate, and an emulsifier, namely potassium soap. The sucrose and fatty acid methyl ester reactants, the potassium soap emulsifier and the potassium carbonate catalyst were fed to the first reaction zone in the initial reaction stage, i.e., reactor R600, at substantially constant rates, and the operating parameters of reactor R600 were maintained substantially constant. Additionally, during the same time period, the amounts of the respective sucrose esters, the amount of unreacted sucrose, the amount of unreacted fatty acid methyl ester, the amount of methanol by-product, the amount of potassium soap emulsifier and the amount of potassium carbonate catalyst leaving reactor R600 were substantially constant. Accordingly, the reaction mixture in R600 was in steady state with continuous introduction of reactants, continuous removal of products and overall mass-balanced in-going reactant and out-going product streams. Additionally, the out-going product stream which was continuously removed had a polyol conversion of more than 1% and lower alkyl alcohol (methanol) was formed during the initial stage of the synthesis and was removed therefrom.

22. As noted above and as set forth in the Hardy and Hawkins Declarations, Exhibit 8 comprises accurate copies of data sheets which set forth determined area percentages of respective esters in the sucrose ester in reactor samples. Exhibit 8 includes data sheets setting forth determined area percentages of octaester in recirculation loop samples from the fourth, fifth

and sixth reactors, R603, R604 and R605, respectively, during the P90117 pilot plant continuous process for the time period of from 4:00 a.m. to 13:00 (1:00 p.m.). More specifically, the area percentages of octaester set forth in Exhibit 8 were as follows:

Table 8: Area Percentage of Octaester in Sucrose Esters in R603, R604 and R605 Samples on January 20, 1989

Time	R603	R604	R605
4:00	68.4	86.9	n/a*
5:00	n/a	n/a	92.4
6:00	67.3	85.3	n/a
7:00	n/a	n/a	94.08
8:00	65.9	86.06	n/a
9:00	n/a	n/a	93.78
10:00	65.61	86.46	n/a
11:00	n/a	n/a	92.95
12:00	67.73	83.60	n/a
13:00	n/a	n/a	95.49

*n/a = data not available

As the samples from the recirculation loop are representative of the out-going product stream, Exhibit 8 demonstrates that octaester was present in the sucrose ester products in the R603, R604 and R605 out-going streams for the noted period of time during the P90117 pilot plant continuous process. Since no octaester was present in the out-going streams from the first reactor R600 as discussed in ¶15, Exhibit 8 demonstrates that the reaction mixture in the out-going product stream from reactor R600 was further reacted to the polyol fatty acid esters (sucrose octaesters) in subsequent reaction stages in at least reactors R603-R605.

23. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Respectfully submitted,

By: Scott D. Pearson
SCOTT D. PEARSON

Date: 4/5/99

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